

Cancer Risk of Inhalation Exposure to Cd, Cr, As, Be and Ni in Ambient Air

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Abstract

Particulate matter may comprise toxic trace elements with carcinogenic effects. Of these trace elements, Cadmium (Cd), Chromium (Cr), Arsenic (As), Beryllium (Be) and Nickel (Ni) are classified as probable human carcinogens. During the annual Hajj pilgrimage, 2.3 million pilgrims stay in Makkah, Saudi Arabia; the increased vehicle traffic contributes to elevated concentrations of particulate matter. We aimed to determine the excess cancer risk (ECR) associated with inhalation exposure in the Makkah population. This study was conducted in the Arafat area, which is a highly-crowded area during Hajj. Inductively coupled plasma-mass spectrometry (ICP-MS) was used for trace element analysis. ECR resulting from inhalation exposure to each metal was calculated in accordance with the unit risk suggested by the Integrated Risk Information System. In summer, including Hajj, mean Cd, Cr, As, Be, and Ni atmospheric concentrations were 0.098, 0.008, 0.26, 0.03, and 0.012 $\mu\text{g}/\text{m}^3$, respectively; in autumn, values were 0.06, 0.006, 0.16, 0.002, and 0.01 $\mu\text{g}/\text{m}^3$, respectively. ECRs were 1.08×10^{-4} , 7.21×10^{-4} , 4.0×10^{-6} , 4.6×10^{-6} , and 2.4×10^{-6} , respectively, exceeding the acceptable inhalation risk level (1.0×10^{-6}) set by the US Environmental Protection Agency for each element. Higher atmospheric trace element concentrations in summer were due to high temperatures that increased atmospheric turbulence, leading to a greater amount of re-suspended dust from roads and blowing sand particles. These findings can be used by relevant authorities while developing regulations and strategies for developing air quality management to improve the health of pilgrims; however, larger prospective studies are required to estimate ECR in different seasons.

Keywords

Carcinogens, Trace Elements, Particulate Matter, PM_{10}

1. Introduction

Since the metals can exist in a particle form, they can contribute to particulate matter (PM) levels or react with gases in the atmosphere to form pollutant compounds [1]. Atmospheric PM is more complex than other air contaminants because of its chemical composition and physical properties, including density, concentration, and size distribution [2]. PM may consist of several toxic trace heavy metals that are potentially harmful to human health because of their carcinogenic properties [3] [4] [5] [6] [7].

The United States Environmental Protection Agency (USEPA) considered Cd as a potential human carcinogen and had classified it as a group B1 carcinogen. The inhalation risk unit (the estimated increased cancer risk from inhalation exposure to a concentration of $1 \mu\text{g}/\text{m}^3$ over an individual's lifetime) of Cd has been calculated as 1.8×10^{-3} ($\mu\text{g}/\text{m}^3$). USEPA estimated that, if a person were to continuously breathe air containing Cd at a concentration of $0.0006 \mu\text{g}/\text{m}^3$ ($6 \times 10^{-7} \text{mg}/\text{m}^3$) throughout his or her lifetime, he or she would theoretically have less than one in a million risk of cancer development due to Cd inhalation. In contrast, those continuously breathing air containing $0.06 \mu\text{g}/\text{m}^3$ ($6 \times 10^{-5} \text{mg}/\text{m}^3$) Cd would have an approximately one in 10,000 risk of cancer development due to Cd inhalation [8].

Chromium is classified as a Group A carcinogen. A previous occupational study of chromate production workers reported that continuously breathing Cr-contaminated air was associated with an inhalation risk unit for Cr of 1.2×10^{-2} ($\mu\text{g}/\text{m}^3$)⁻¹. In addition, those who continuously breathe Cr-contaminated air ($0.00008 \mu\text{g}/\text{m}^3$ [$8 \times 10^{-8} \text{mg}/\text{m}^3$]) throughout their lives may have less than one in a million risk of cancer [8] due to Cr inhalation. Arsenic is also classified as a group A carcinogen. Studies have reported a strong association between As inhalation and lung cancer risk. Moreover, studies have reported a strong association between As ingestion or exposure and elevated risk of bladder, liver, lung, and skin cancers [9] [10]. Beryllium is a group B1 carcinogen. Studies have reported a causal relationship between Be exposure and lung cancer [11] [12]. Nickel dust and Ni bisulfide are categorized by the USEPA as group A carcinogens. Ni carbonate, Ni carbonyl, Ni hydroxide, and Ni sulfate are classified by the US National Occupational Health and Safety Commission as category 3 carcinogens and Ni oxide and Ni bisulfide as category 1 carcinogens. In addition, Ni carbonyl is classified as a category 2 carcinogen [13].

Some previous studies indicated that PM_{10} levels in Makkah, Saudi Arabia exceeded the national and international standards set for the protection of human health [14] and suggested a detailed investigation of the effects of PM_{10} and $\text{PM}_{2.5}$ exposure on human health using data from multiple locations [15] [16]. Another study revealed high levels of PM_{10} in different location in Makkah (Al Haram) [17]. Moreover, previous study in Makkah showed significantly positive trends in PM_{10} concentrations; several reasons for the high particulate concentrations in Makkah were suggested, including an increasing number of diesel vehicles, especially during the Hajj season for transportation of pilgrims, increasing construc-

tion activities in Makkah, changing weather patterns, and increasing proportion of windblown and re-suspended dust particles [18].

Hence, the present study aimed to determine the ECR associated with inhalation exposure to five heavy metals (Cd, Cr, As, Be, and Ni) in ambient air. Since the carcinogenicity risk for trace elements in Makkah is unknown, ECR from inhalation exposure to each metal was calculated. To our knowledge, no modeling studies have been published concerning air pollution in Makkah and its expected effect on population health, especially regarding cancer risk. Thus, this is the first study to examine cancer risk with respect to exposure to trace elements in Makkah.

2. Materials and Methods

2.1. Sampling Location

This study was conducted in the Arafat area (21.35 [21°21'1"N] latitude; 39.97 [39°58'1"E] longitude) east of Makkah, **Figure 1**. The Arafat area was selected as it represents a highly-crowded area during the Hajj season, **Table 1** and **Table 2** [19].

2.2. Meteorological Measurements

On-site measurement of air temperature, wind speed, wind direction, ultraviolet (UV) radiation, and rainfall rate was performed using the Davis Instruments 6163 Vantage Pro2 Plus (Davis Instruments, USA). Meteorological data were recorded at a height of 10 m on a 47-mm Teflon filter at 16.6 l/min flow rate for 24 hours once a week for 6 months during the summer and autumn in 2014, in accordance with the USEPA standard method (Method 29 as updated in 2000). Weather information was collected, including data regarding temperature, humidity, rainfall amount, wind speed, wind direction, wind chill factor, and barometric pressure. UV radiation was determined by measuring the irradiance at UV wavelength, as

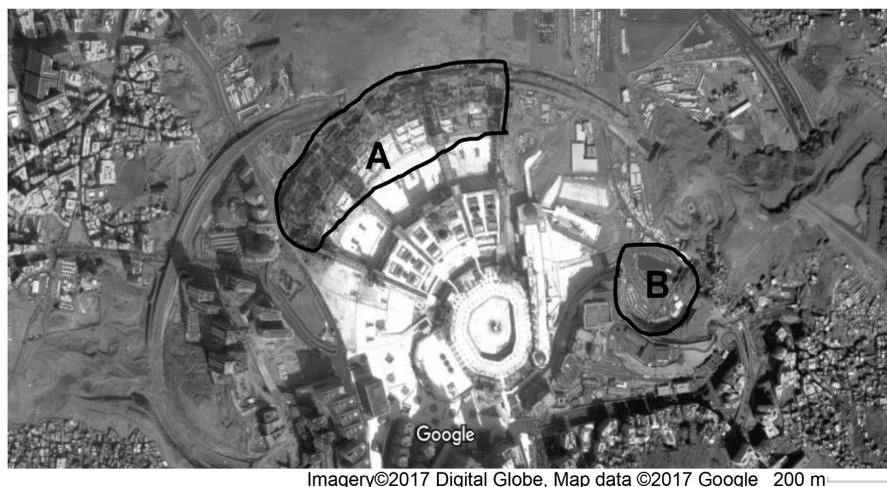


Figure 1. Map of the Makkah area showing various sources of emissions. (A) shows the construction and development area of Masjid Al-Haram; (B), Main bus station stand along Masjid Al-Haram.

Table 1. Number of pilgrims from 1995-2012 [19].

The number of pilgrims for the years from 1416H (1995G) to 1433H (2012G)			
Year (H/G)	Pilgrims from Outside Saudi Arabia	Pilgrims from Saudi Arabia	Total
1416/1995	1,080,465	784,769	1,865,234
1417/1996	1,168,591	774,260	1,942,851
1418/1997	1,132,344	699,770	1,832,114
1419/1998	1,056,730	775,268	1,831,998
1420/1999	1,267,555	571,599	1,839,154
1421/2000	1,363,992	549,271	1,913,263
1422/2001	1,354,184	590,576	1,944,760
1423/2002	1,431,012	610,117	2,041,129
1424/2003	1,419,706	592,368	2,012,074
1425/2004	1,534,769	629,710	2,164,479
1426/2005	1,557,447	700,603	2,258,050
1427/2006	1,654,407	724,229	2,378,636
1428/2007	1,707,814	746,511	2,454,325
1429/2008	1,729,841	679,008	2,408,849
1430/2009	1,613,965	699,313	2,313,278
1431/2010	1,799,601	989,798	2,789,399
1432/2011	1,828,195	1,099,522	2,927,717
1433/2012	1,752,933	1,408,641	3,161,573

H, Hijri calendar; G, Gregorian calendar.

Table 2. Number of vehicles carrying pilgrims in Makkah, per car type [19].

Vehicle Type	Numbers of Vehicles		Difference	Percentage (%)
	1432H/2011G	1433H/2012G		
Small	39,846	43,482	+3636	91
Salon	16,277	17,580	+1303	80
Pick-up truck	4595	5732	+1137	247
Jeep	8842	9342	+500	57
Small bus	5947	6077	+130	22
Big small	8325	12,166	+3841	461
Trucks	231	366	+135	584
Others	4915	4699	-216	-44
Total	88,978	99,444	10,466	118

H, Hijri calendar; G, Gregorian calendar.

the value significantly varies in ozone and cloud cover. Data (1-min averages) were automatically stored in a data log every 15 min for 1 year; daily averages were calculated. All data were sent directly to the laboratory via a modem. In all cases, data were collected in strict compliance with standards regarding meteorological monitoring equipment [20].

2.3. PM₁₀ Sampling and Analysis

24 air samples were collected during summer, including the Hajj pilgrimage, and autumn using a mini volume sampler (Airmetrics, USA) at a height of 10 m on a 47-mm Teflon filter at 16.6 l/min flow rate for 24 hours once a week for 6 months, in accordance with the USEPA standard method (Method 29/2000). PM samples were collected at a height of 10 m in the Arafat area on a filter of mixed cellulose ester membrane with a diameter of 47 mm (Whatman, Grade 1 Qualitative Filter Paper Standard Grade, USA). Each filter with the collected particles was placed and stored flat on a clean Petri dish during and after conditioning for weighing and storage. Filters were weighed pre- and post-sampling using an electronic microbalance (CITIZEN Micro Balance Model CM21P, USA). The filters were conditioned in a dry-keeper before and after collection. Teflon filters for PM₁₀ were weighed and conditioned at a temperature of 35°C - 40°C and humidity of 60% - 70% ± 5%.

Sample filters were collected weekly for individual analysis. Particles collected on each filter were extracted with 7 ml of nitric acid and 2 ml of ultra-pure water (ASTM type 1 water from Millipore filtration system, Millipore Cooperation, Massachusetts, USA) followed by microwave-assisted acid digestion using 5 ml of concentrated nitric acid, 3.0 ml of concentrated hydrofluoric acid, 2.0 ml of concentrated hydrochloric acid, and 1.0 ml of hydrogen peroxide in each sample vessel. The sample vessels were sealed and placed in a rotor (8 × 100) for microwaving. Samples were analyzed for Cd, Cr, As, Be, and Ni concentrations in triplicate using inductively coupled plasma-mass spectrometry with a Perkin Elmer 7300 (Perkin Elmer, USA) per the manufacturer's instructions. Trace elements were detected using the appropriate wavelengths as per USEPA Method 200.7, and two factors were considered: The freedom from spectral interferences and the different sensitivities against the expected sample concentration. The observed interferences were compensated for by modifying the processing parameters, which was accomplished by adjusting the background correction points **Table 3**.

2.4. Quality Assurance and Quality Control Procedures

Quality assurance and control (QA/QC) were performed for each sample by analyzing a control sample regularly with the samples to ensure reliability; reproducibility and linearity were determined for each analysis. A linear calibration curve was constructed using a blank and a five-point calibration curve using the following concentrations: 0.01, 0.1, 0.2, 0.5, and 1.0 ppm for each of the five elements

Table 3. Instrumental and data acquisition parameters of the ICP-Perkin Elmer 7300.

Instrumental parameters		Data acquisition	
RF power	1400 W	Measuring mode	Segmented scan
Argon gas flow	13 - 16 l/min	Point per peak	5
Nebulizer	1.0 l/min	Scans/replicates	6
Plasma	18.0 l/min	Replicate/sample	6
Sample uptake rate	190 s	Integration time	398.6 s

standards. All QC samples and samples containing elements with measured concentrations were within the range of the calibration curve. All calibration samples, QC samples, and production samples were analyzed using 2% nitric acid. In all 0.001-ppm QC checks, the determined concentration was within 20% of the true value, and the relative standard deviation was less than 6%.

2.5. Determination of ECR

ECR was measured in accordance with the unit risk suggested by the Integrated Risk Information System based on the inhalation exposure to each metal for the five heavy metals in ambient air [21]. ECR for each metal was determined using the following mathematical formula: $ECR = C_{\text{pollutant}} \times IUR$, where $C_{\text{pollutant}}$ refers to the mean concentration of heavy metal (μg) and IUR refers to the inhalation unit risk for each metal, as determined using risk assessment data from the USEPA. The IUR for a human group B1 carcinogen is defined by the USEPA as the estimated upper limit of lifetime ECR resulting from continuous exposure to an agent at a concentration of $1 \mu\text{g}/\text{m}^3$ in ambient air. Therefore, the IUR is the greatest level of ECR for either mesothelioma or lung cancer from chronic inhalation exposure in the general US population. IURs are based on data obtained from epidemiologic studies on humans. The approach to determine the IUR from human epidemiologic data involves the quantitative evaluation of the exposure-response relationship (slope) for each element in the studied population [22].

The monthly mean concentration of trace elements was grouped for temporal comparisons to determine whether there was a statistical difference in the mean values per season. The two seasons included in the study were autumn (September through November) and summer (June through August). To test the null hypothesis that there would be a difference in the mean concentration of trace elements according to season, one-way ANOVA (SPSS, 2007) was performed with a significance level of $\alpha = 0.05$.

3. Results and Discussion

Table 4 shows the descriptive statistical analysis of PM_{10} concentrations in the Arafat area in Makkah during the summer and autumn seasons. The average mass concentration of PM_{10} was 1.5 times higher in summer. This higher particulate concentration in summer may be attributed to re-suspension of dust from

Table 4. Twenty-four-hour average particulate matter (PM_{10}) concentrations ($\mu\text{g}/\text{m}^3$) in the Arafat area from June to November 2014.

Months	Mean	SD	Min	Max
June	240.1	33.8	100.7	498.7
July	223.4	45.3	122.7	830.3
August	167.8	56.9	108.8	263.7
September	168.7	39.4	121.5	308.8
October	190.6	30.7	96.7	330.2
November	171.7	37.7	102.9	218.6

SD, Standard deviation.

roads, natural dust storms, and automobile traffic. PM_{10} concentrations in the Arafat area increased during the pilgrimage period in summer owing to the high increase in pollution problems due to transportation, ultimately resulting in an unspecified amount of trace elements causing air pollution. Mean concentrations of airborne trace elements were significant per season.

A previous study, conducted in Makkah, reported that the annual average PM_{10} level was $233.38 \mu\text{g}/\text{m}^3$ [18], whereas a 2012 study in Jeddah showed an annual average PM_{10} concentration of $87.3 \mu\text{g}/\text{m}^3$ [13]; this is expected as Jeddah and Makkah have different pollution sources and atmospheric conditions. The weekly average PM_{10} concentration was highest on weekdays and lower on weekends [15]. In the present study, the highest monthly PM_{10} concentrations were observed in June and July (summer including Hajj); the corresponding concentrations were 240.1 and $223.4 \mu\text{g}/\text{m}^3$, respectively, which was significantly correlated with seasonal variation and the increased transportation and traffic activities during Hajj ($P < 0.05$); this has also been reported for the summer season by other studies which did not include Hajj [22]. The present study showed that a high PM concentration is strongly associated with the nature of the region, including temperature, wind speed and direction, humidity, and number of vehicles.

Regarding the trace element concentrations, the mean atmospheric concentrations of Cd, Cr, As, Be, and Ni were 0.098 , 0.008 , 0.26 , 0.03 , and $0.012 \mu\text{g}/\text{m}^3$, respectively, in summer, while the corresponding values in autumn were 0.06 , 0.006 , 0.16 , 0.002 , and $0.01 \mu\text{g}/\text{m}^3$, respectively, **Figure 2**. Among the samples of PM_{10} in the Arafat area, As concentration was the highest in all studied months in different seasons, **Figure 3**; this is strongly aligned with the results of another study conducted in Makkah that reported that As, Hg, and Al had the highest concentrations in different samples of PM_{10} , total suspended particles, and $PM_{2.5}$ [23]. The higher heavy metal concentration in summer is likely due to high temperature inversion, during which an increased number of PM_{10} particles are found closer to the surface owing to increased atmospheric turbulence and blowing dust, thus metal contaminants were transported and dispersed in the surrounding areas. The high concentration of As, a group A carcinogen, may cause a serious health threat to the Makkah population.

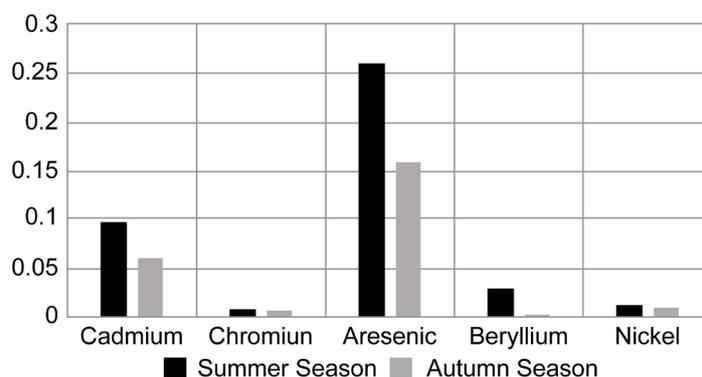


Figure 2. Seasonal average concentrations of trace elements in PM_{10} at the monitoring site in Arafat area from June to November 2014.

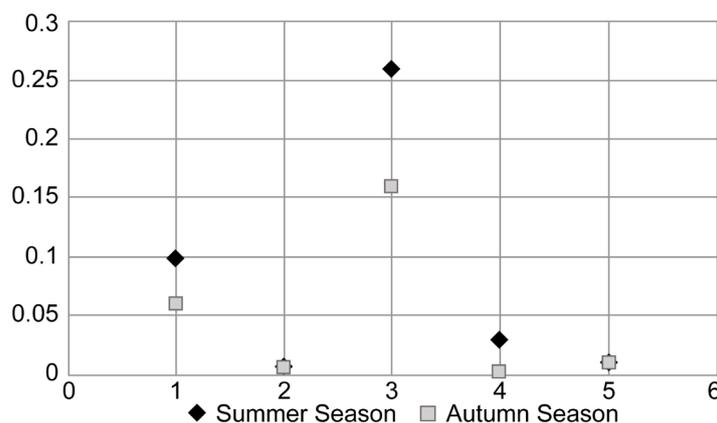


Figure 3. Correlation plot of trace elements in PM₁₀ at the monitoring site in Arafat area from June to November 2014.

3.1. Meteorological Measurements

The highest average wind speed of 4 m/s (gentle breeze) was noted in June, at which time the average daily maximum wind speed was 8 m/s (fresh breeze). The lowest average wind speed of 3 m/s (light breeze) was noted in October, **Figure 4**. All other meteorological parameters measurements were in the seasonal normal range.

3.2. Risk Assessment

The ECRs for the Arafat area were 1.08×10^{-4} , 7.21×10^{-4} , 4.0×10^{-6} , 4.6×10^{-6} , and 2.4×10^{-6} for Cd, Cr, As, Be, and Ni respectively, **Table 5**; these values exceeded the level of acceptable inhalation risk (1.0×10^{-6}) for each element set by the USEPA [8].

4. Conclusions

Arsenic had the highest concentration among all the five carcinogenic trace elements studied. The ECRs for the Arafat area for each of the five trace elements exceeded the level of acceptable inhalation risk (1.0×10^{-6}) for each element set by the USEPA. Although it is not possible to accurately determine the population exposure to ambient air pollution as there is limited knowledge regarding time-activity patterns, pollutant concentrations are considered to indicate the exposure level.

To the best of our knowledge, this is the first study to assess airborne trace element concentrations and their correlation with cancer risk in Makkah. Further studies are required to monitor and assess additional trace elements and their carcinogenic effect as well as the effects of long- and short-term exposure and the contribution of different air pollution sources, particularly road traffic, which is the main source of several air pollutants in urban areas. Moreover, larger prospective studies are warranted to explore the health effects of long-term exposure to ambient air trace elements. Further monitoring plans should be adopted in Makkah for preventing long-term exposure at different monitoring locations, including pilgrimage sites, roadside, and urban sites.

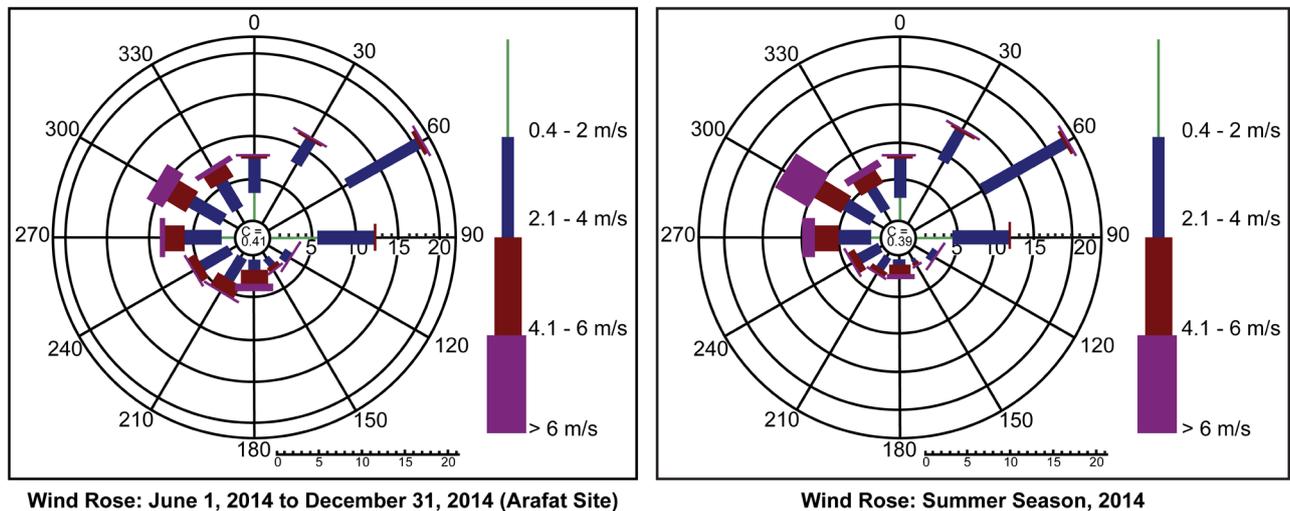


Figure 4. Wind roses for the Arafat area during the 2014 fall and summer seasons showing wind direction and the percentage of time that winds blew from a direction at certain speed ranges. Wind speeds shown in the plots are in m/s.

Table 5. Assessment of cancer risk in the exposed population based on trace element concentration in inhalable ambient air from June to November 2014.

Trace Elements	Ambient Exposure ($\mu\text{g}/\text{m}^3$)	Inhalation Unit Risk ($\mu\text{g}/\text{m}^3$)	ECR ($\mu\text{g}/\text{m}^3$)
Cd	0.098	1.8×10^{-3}	1.08×10^{-4}
Cr	0.008	1.2×10^{-2}	7.21×10^{-4}
As	0.016	1.6×10^{-3}	4.4×10^{-6}
Be	0.03	4.0×10^{-4}	4.6×10^{-6}
Ni	0.012	2.0×10^{-2}	2.4×10^{-6}

Excess cancer risk.

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Authors' Contributions

All authors equally contributed in the article. All authors read and approved the final manuscript.

Conflict of Interests

The authors declare that they have no competing interests regarding the publication of this manuscript.

Compliance with Ethical Standards

The study protocol was approved by Ethics Review Board for Human Studies at Faculty of Medicine, Umm Al-Qura University and conformed to the ethical guidelines of the 1975 Helsinki declaration.

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